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Title: Destructive and Non-destructive Assay of Special Nuclear Material

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Destructive and Non-destructive Assay of Special Nuclear Material

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4 May 2021

ETI 101: Fundamentals of Nuclear Science and Engineering for Nonproliferation

Module 4 - Overview of Nuclear Security and Nonproliferation

Destructive and Non-destructive Assay Techniques

by Katrina Koehler and Jacob Stinnett

4 May 2021

Consortium for
ENABLING TECHNOLOGIES & INNOVATION

Outline

- Purpose of Destructive and Nondestructive Assay
- Destructive Assay
- Nondestructive Assay by gamma spectroscopy
- Gamma Spectroscopy R&D

Destructive Analysis

Destructive Chemical Analysis

NDA and DA methods are used to quantify nuclear material (uranium, plutonium, etc.) for nuclear material accounting, for process control, and more

DA is used to establish the elemental content and isotopic composition of nuclear materials

Examples:

- trace impurity analysis in plutonium oxide
- precise plutonium oxide isotopic analysis

A small portion of the bulk material is substantially changed chemically, physically, or both as a part of the measurement process, typically becoming waste, thus the term “destructive” analysis

Measurement Process

1. **Bulk measurement**

    Volume, Weight, Flow

2. **Sampling**

    Random grabs, Core samples

3. **Sample Preparation**

    Dissolution, Separation chemistry

4. **Analytical measurement**

Bulk Measurement of Solids

Examples: oxides, scrap materials, product metal

Problems with recorded weights

- Gain or loss of water
- Surface oxidation

Unstable, changes in mass over time

- Loss of volatile components
- Improper technique
- Poor standards
- Aging of balance

Quality Assurance

Sampling and Sample Preparation

Collection of a representative sample from bulk material?

Prepare sample:

- Resampling of the collected sample
- Dissolution (acid or fusions)
- Separations (solvent extraction or ion exchange)
- Chemical treatments (oxidation state adjustment, acid strength adjustment)
- Other (e.g., reweighing, shaving)

Analytical Measurements - Summary of DA Types/Methods

Gravimetric
- Determination of U

Alpha Spectrometry/Alpha counting - Pu

Reduction-Oxidation (Redox) Titrimetry (examples)
- Davies/Gray Titration of U
- Ceric Titration of Pu
- Ferrous Titration of Pu
- Coulometric Determination of Pu

Spectrophotometry/Fluorometry – U and Pu

Mass Spectrometry
- Impurity Analysis
- U or Pu isotopic composition
- U or Pu concentration determination by Isotope Dilution Mass Spectrometry (IDMS)

Multicollector Mass Spectrometry

Method of choice for determination of isotopic composition

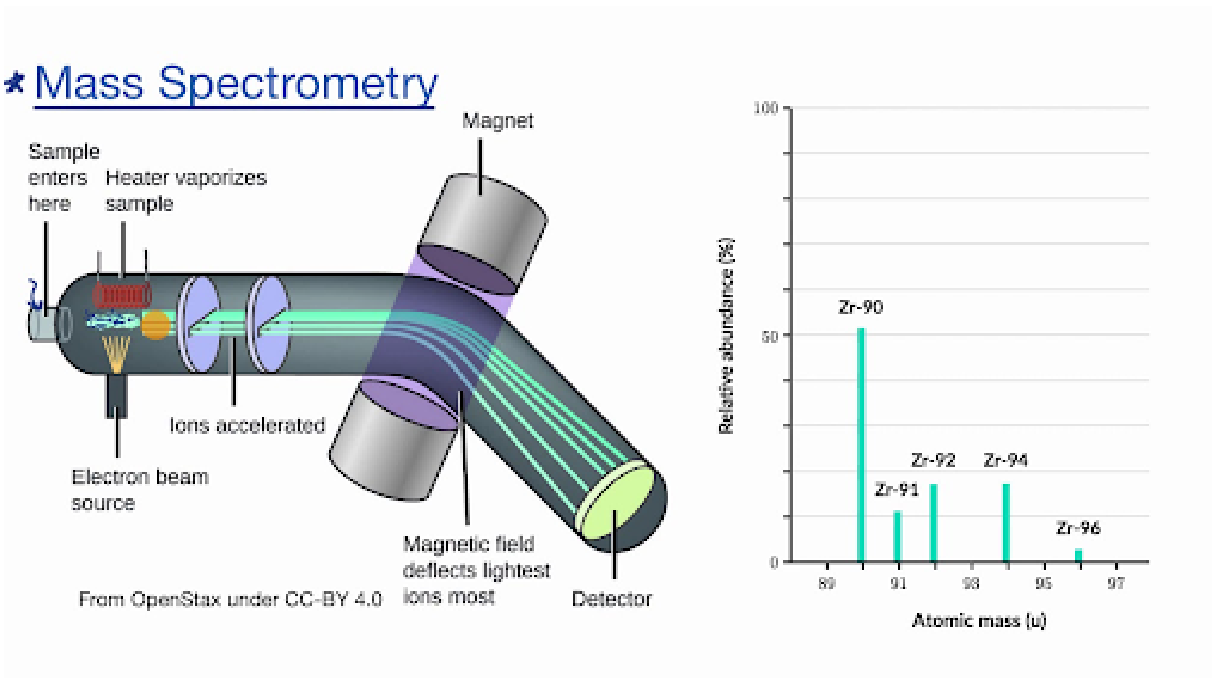
Measures isotopes simultaneously

Very good precision and accuracy

- Precision 0.005-0.1% for U/Pu
- Accuracy 0.002-0.06%

Small samples $10^{-15} – 10^{-5}$ grams

A chemist prepares a sample for an IsotopX Phoenix multicollector thermal ionization mass spectrometer (TIMS)



Chemists working with a Neptune Plus multicollector ICP-MS

Isotope Ratios Measured by Mass Spectrometry

Thermal Ionization Mass Spectrometry (**TIMS**) and Multicollector Inductively Coupled Plasma Mass Spectrometry (**MC-ICP-MS**) applicable to many material types

^{241}Pu vs ^{241}Am

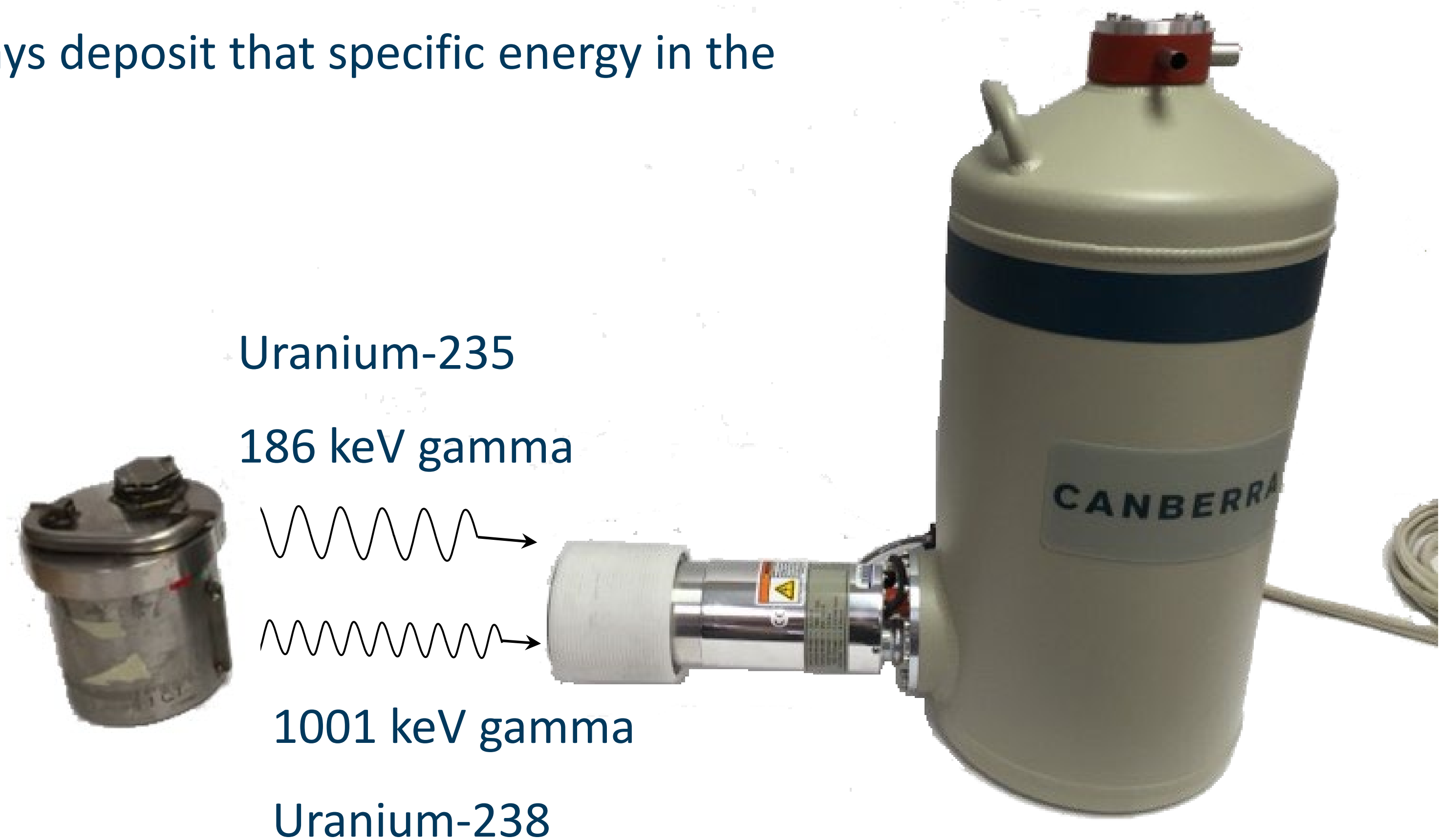
- Isobaric (same mass) interferences must be considered
- Advantages include small sample size and excellent precision
- Disadvantages include complex sample preparation procedures, equipment, and skills

Non-Destructive Analysis: Gamma Spectroscopy

Gamma NDA Summary

Isotopes emit gamma-rays with distinct energies

The gamma-rays deposit that specific energy in the detector

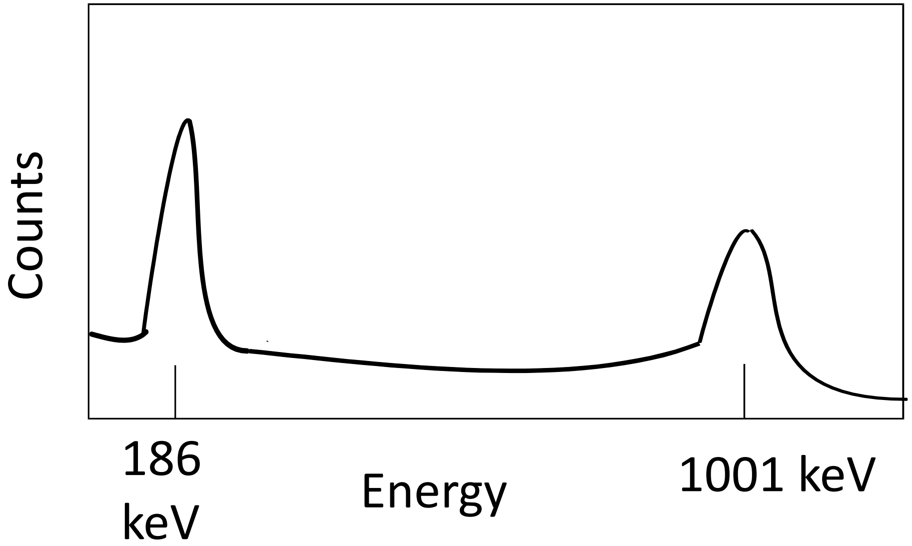


Gamma NDA Summary

When the detector reads a specific energy, we know the corresponding isotope is present

We can tell relatively how much of each isotope is present by the ratio of the counts

- Our item enrichment is 5% ^{235}U → input into MC&A



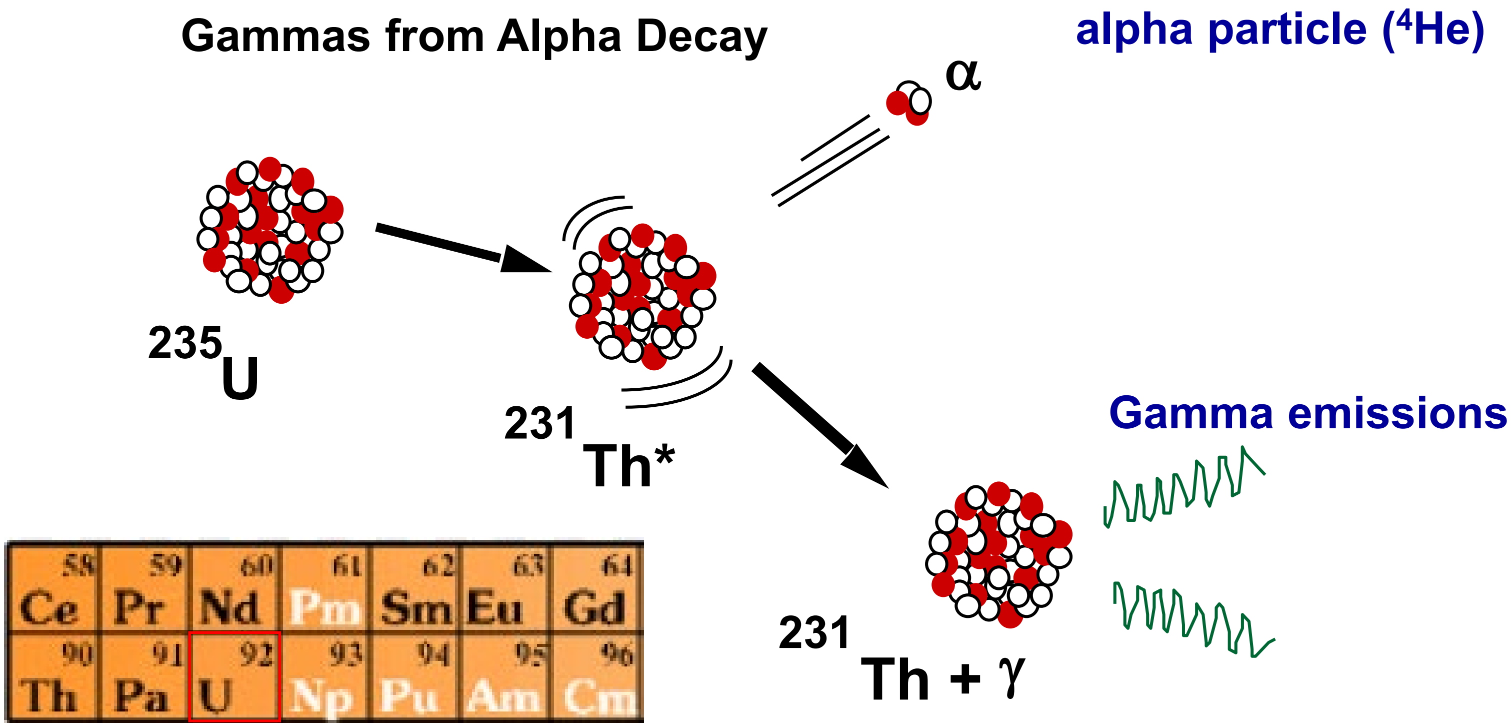
For large items, gamma NDA gives the isotopic composition only. Neutron or calorimetry is needed to find the total mass.

Typical measurement time is 10 minutes, typical uncertainty is 1%

Origin of Gamma-Rays

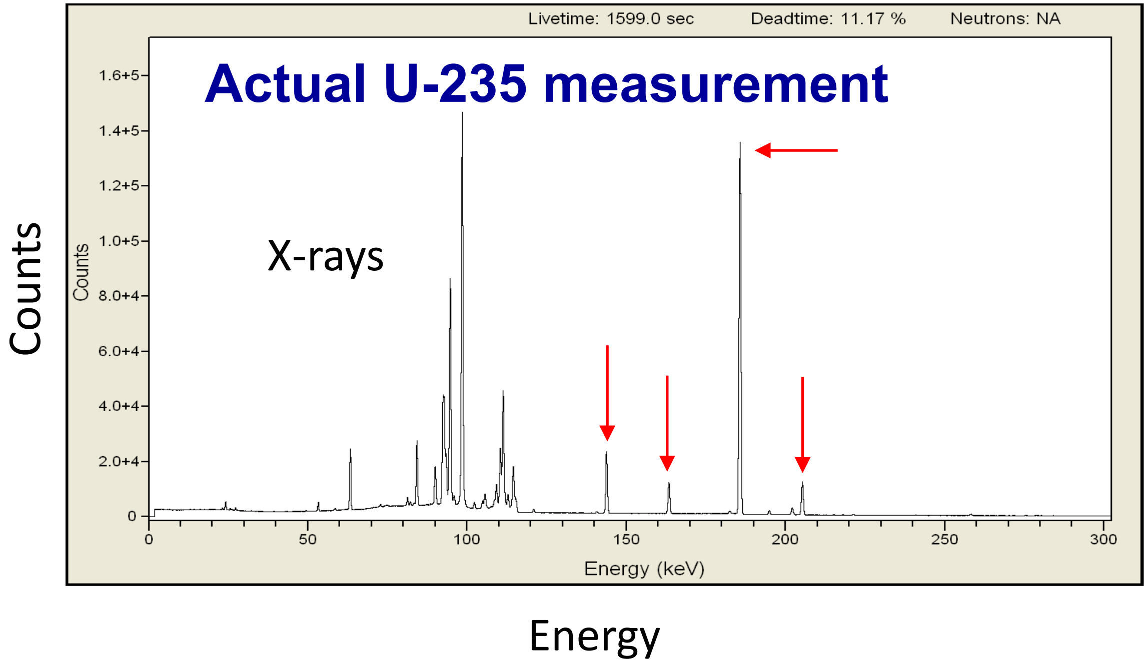
Gammas are emitted from an excited (energized) nucleus

The nucleus de-excites (loses energy) by emitting one or more gamma-rays



Branching Ratios

Different gammas are emitted with a known probability ("branching ratio")

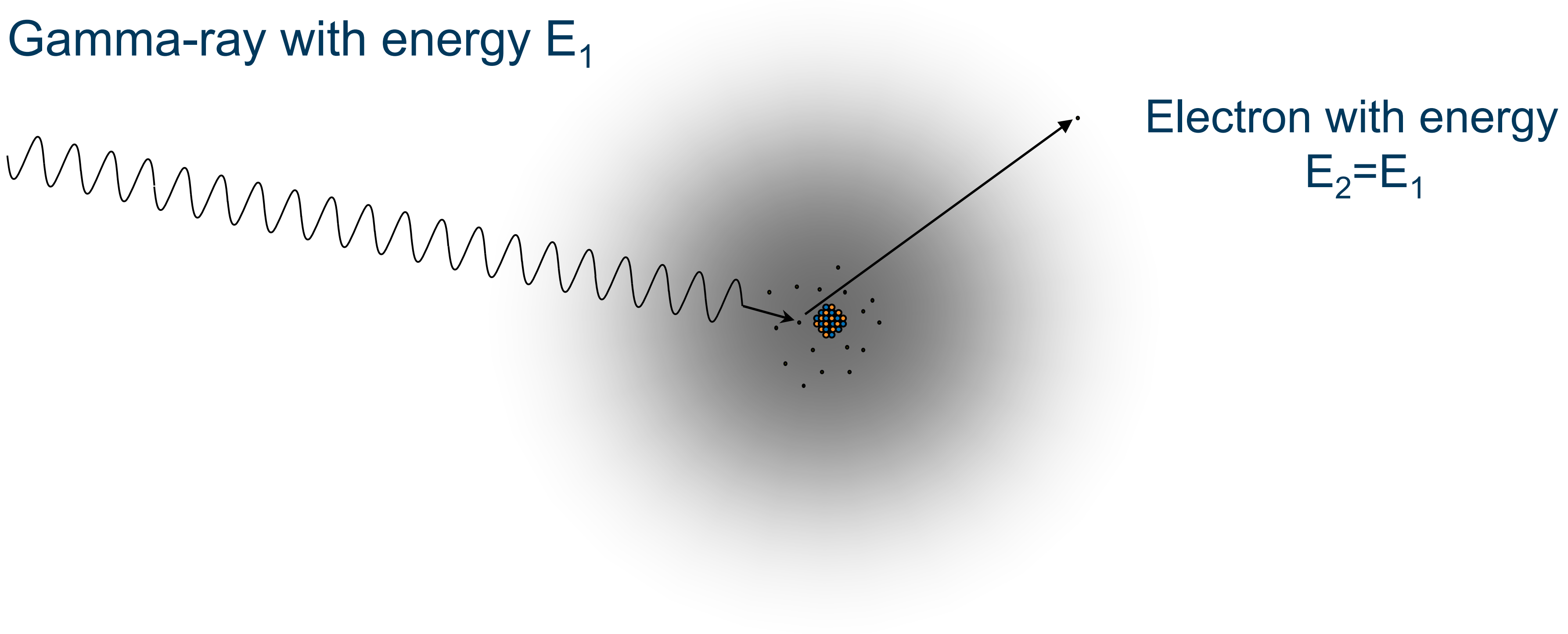


U-235 Gammas:	BR
185.7 keV	57%
144 keV	11%
163 keV	5%
205 keV	5%

The Photoelectric Effect

Gamma-ray fully transfers energy to electron

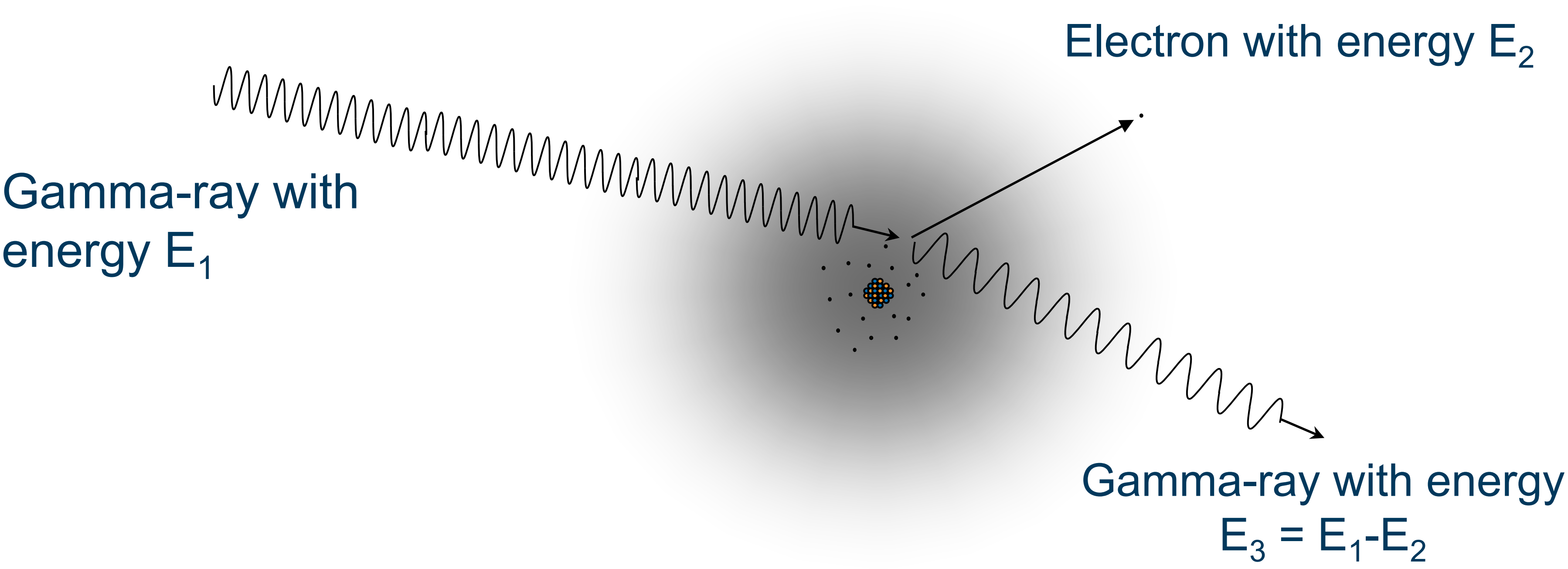
Gamma-ray is **fully absorbed**



Compton Scattering

Gamma-ray transfers some energy to electron

Gamma-ray continues on, **some energy escapes**



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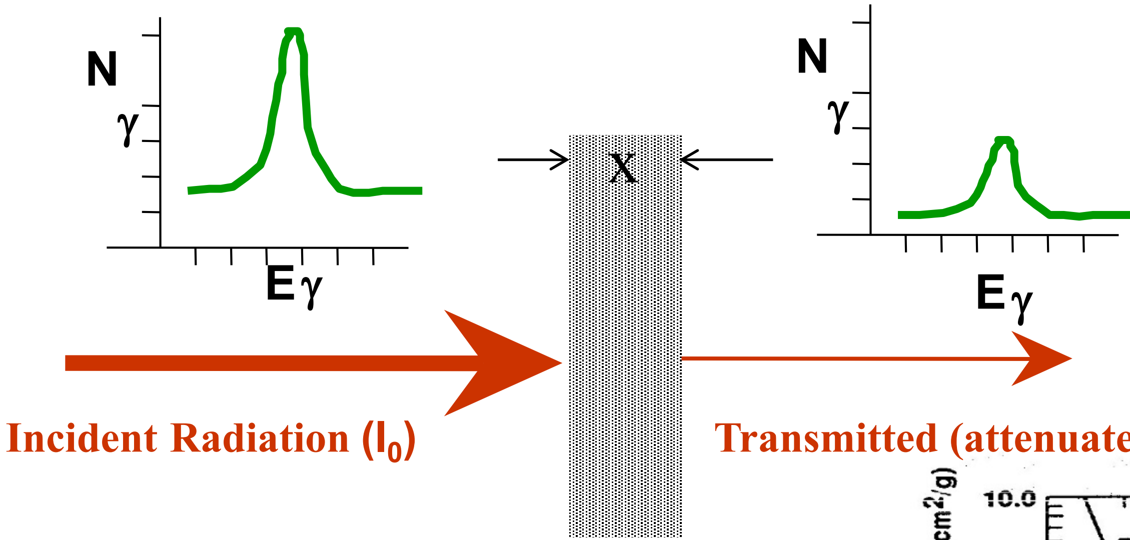
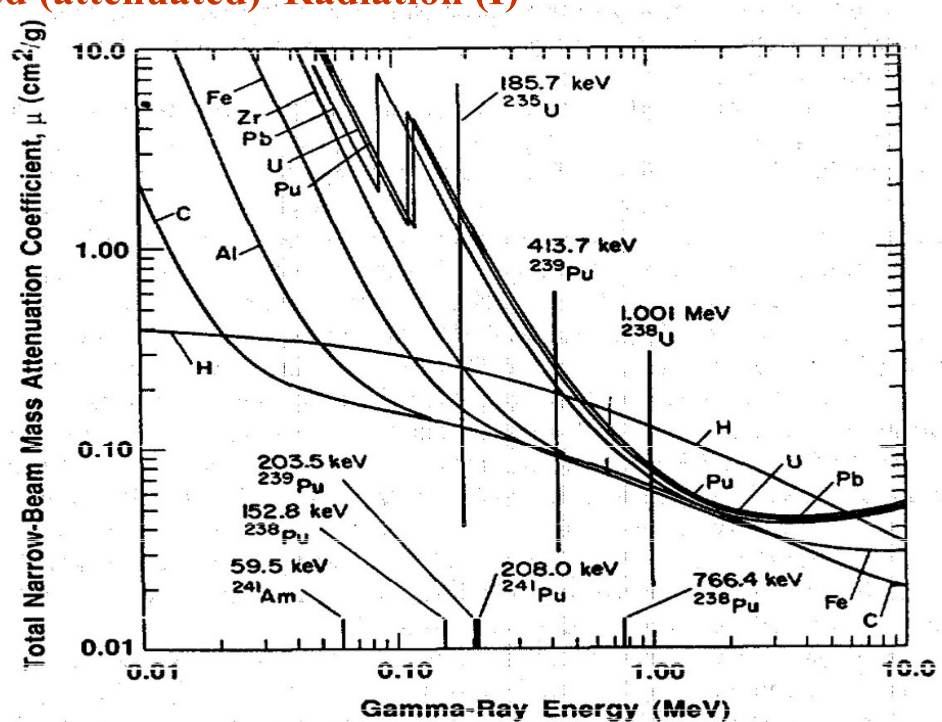
Attenuation of Gamma-rays

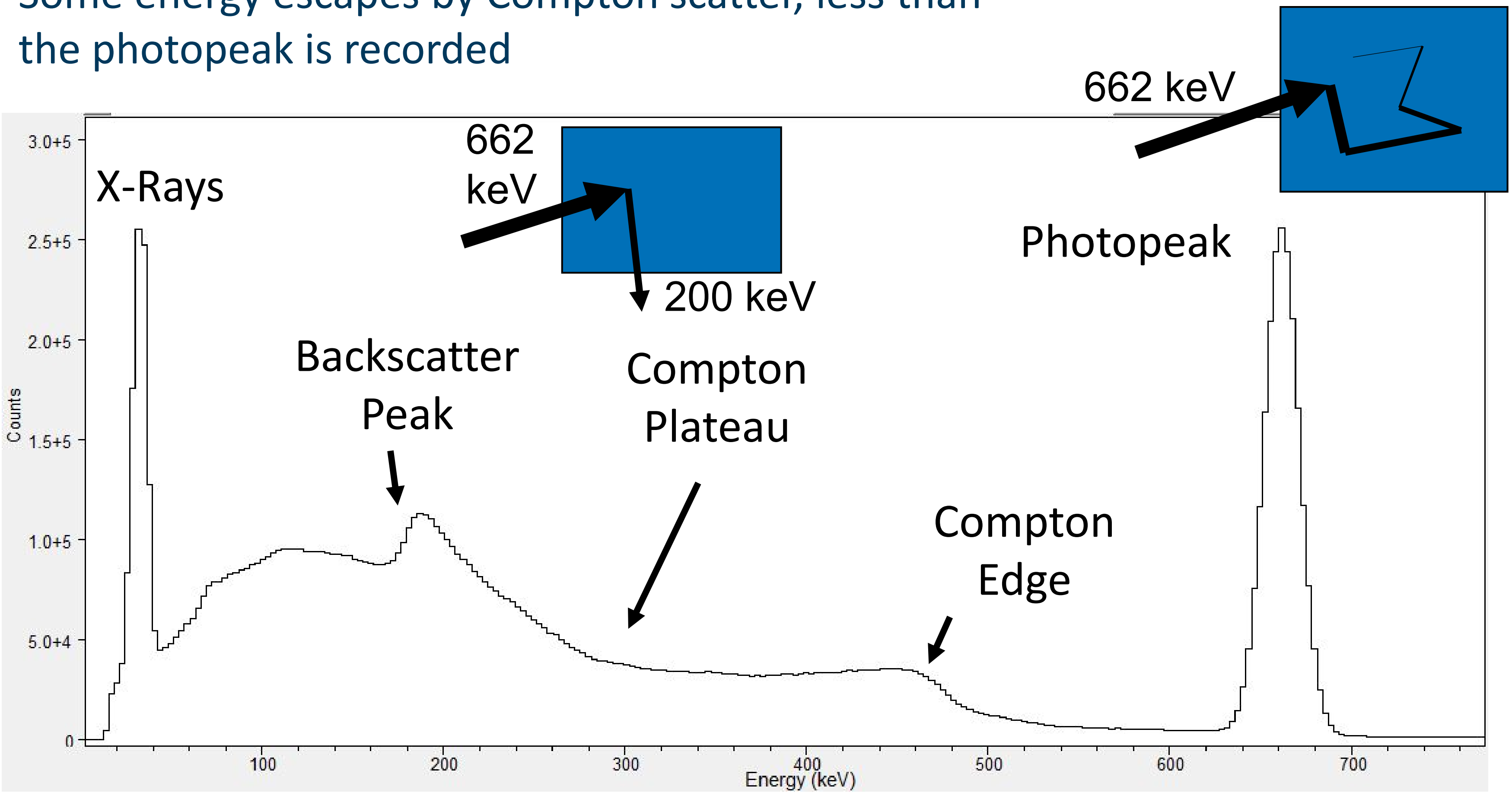


density (g/cm^3)
mass absorption coefficient (cm^2/g)
thickness (cm)



Features of a Monoenergetic Spectrum

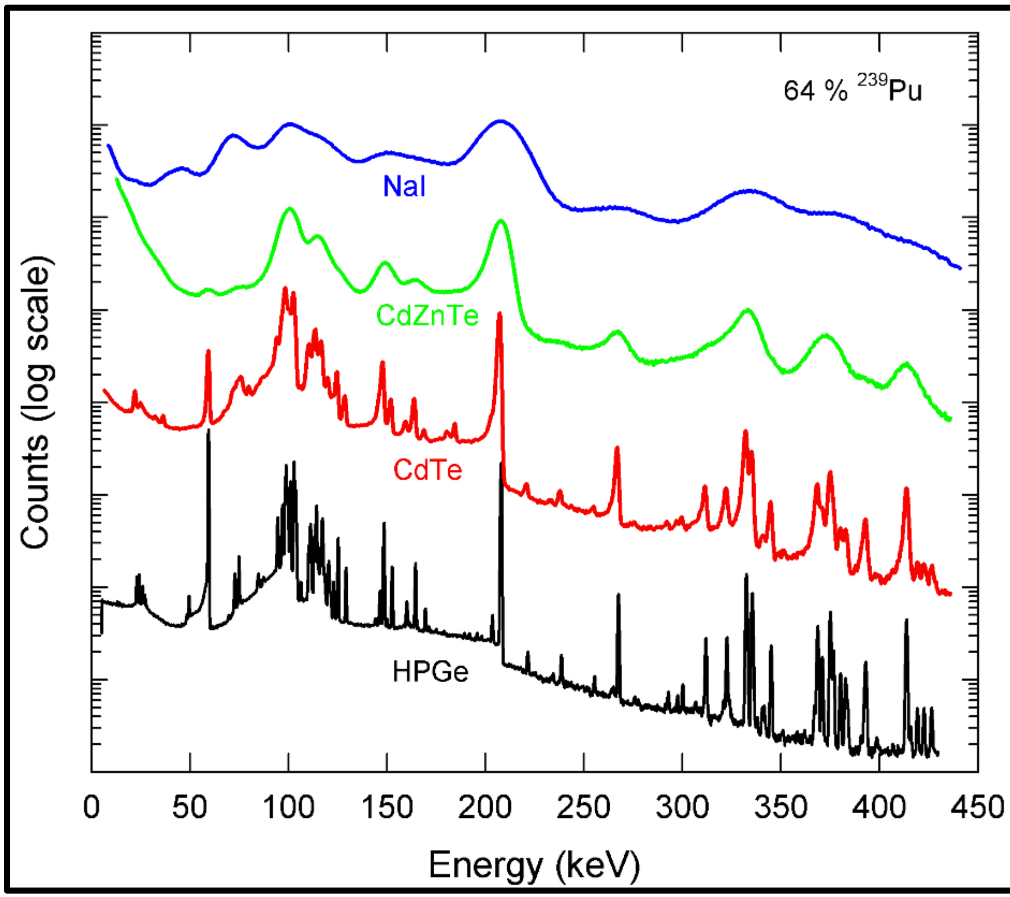
Some energy escapes by Compton scatter, less than the photopeak is recorded



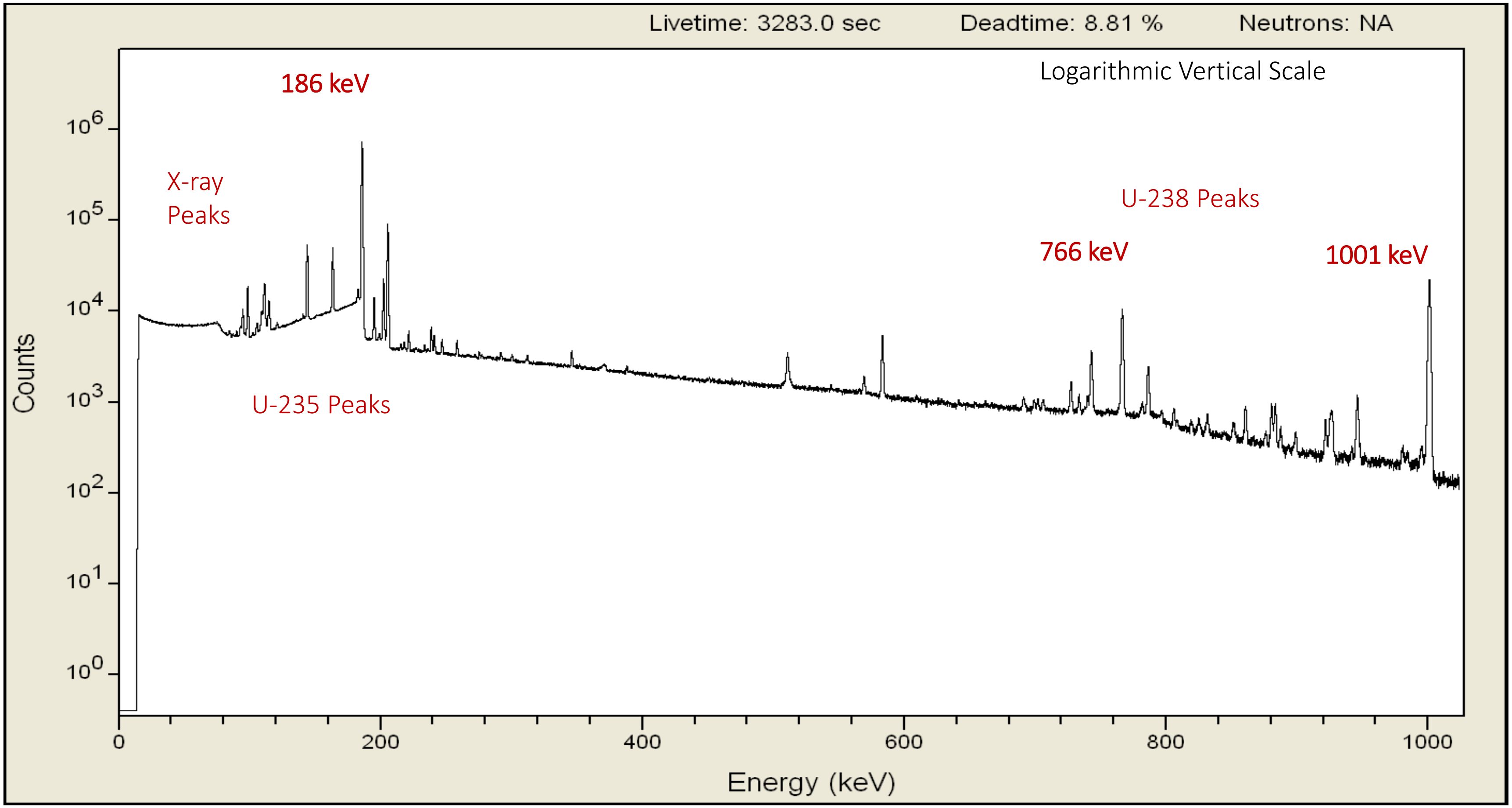
We will mostly focus on full energy peaks

Energy Resolution

Peaks close in energy require good energy resolution in order to be distinguished from each other



Uranium Gamma-Ray Spectrum (1-MeV Range)



Plutonium Gamma-Ray Assay

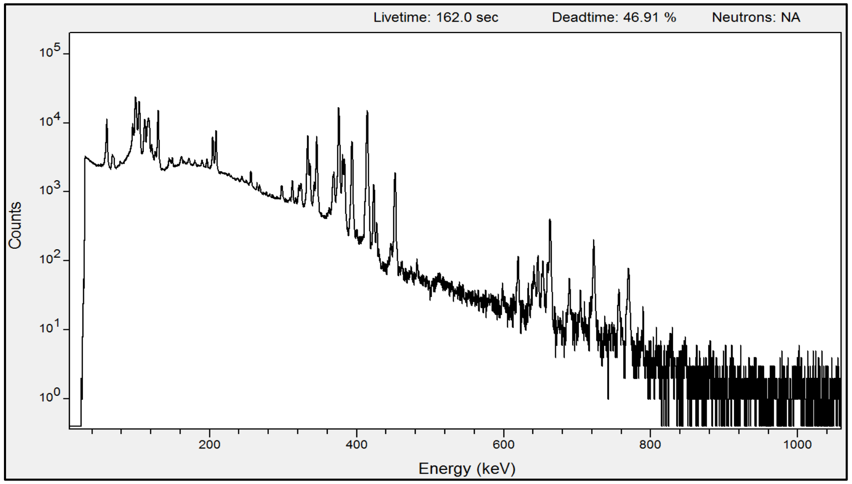
Problem statement

Find the mass of Pu in this container, by isotope

- Measure with Gamma to get isotopic %
- Measure with calorimetry or neutron to get power/effective mass
- Combine for mass of each isotope
- Feed into MC&A needs

1. Measurement

2. Spectrum



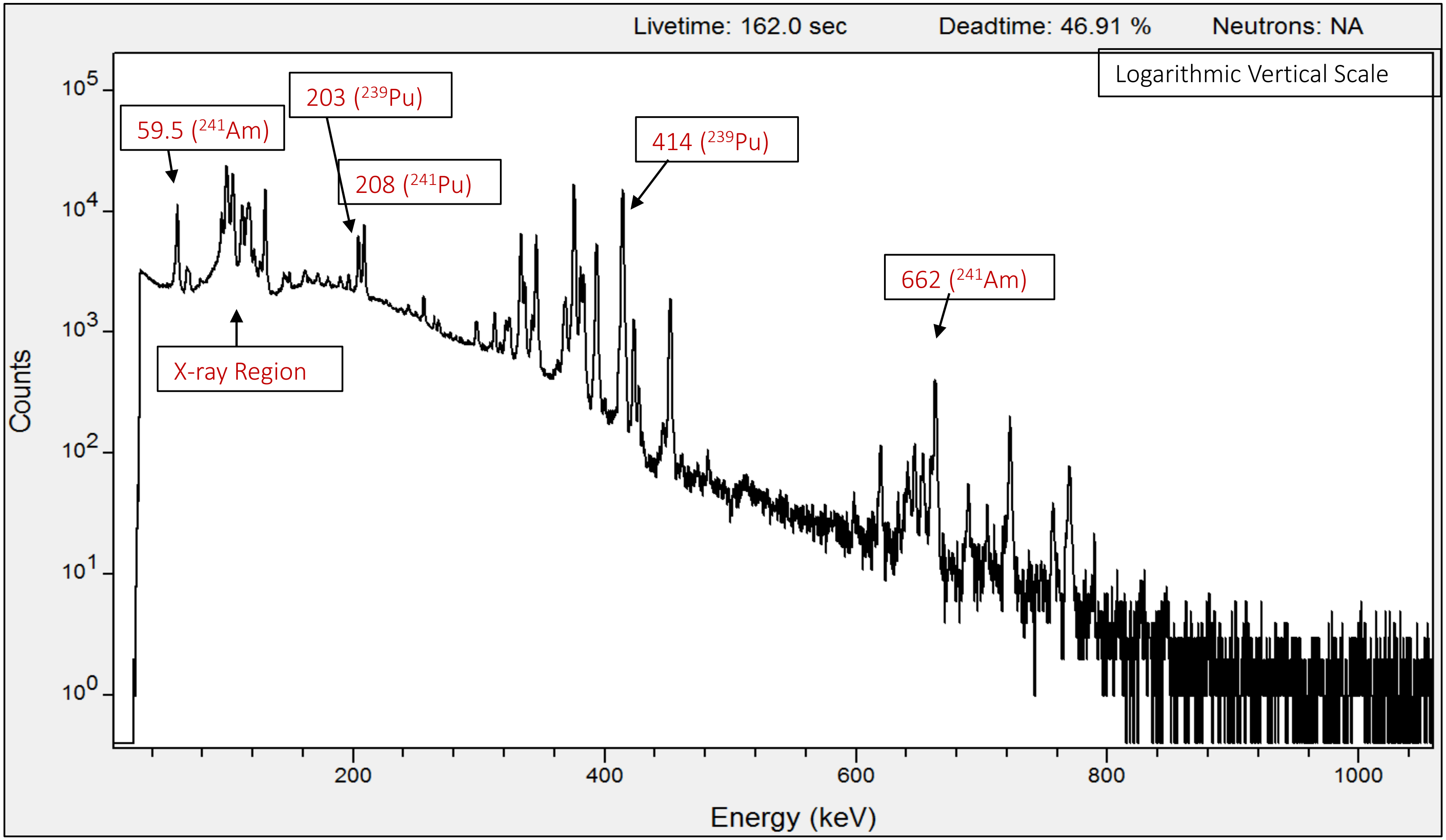
Plutonium Gamma-Ray Assay

3. Isotopic ratio analysis with FRAM
   - Compares how big each peak is to determine isotopics
   - Considers detector efficiency, half-lives, branching ratios, peak shapes, backgrounds, energy calibration, etc.

```
***************************************************************************
PC FRAM (6.1)      Isotopic Analysis      16-Apr-2020 09:07:34
(Fixed energy Response function Analysis with Multiple efficiencies)
Operator ID:

spectrum source:  C:\FRAMdata\Testspec\TestSpec_chn\93coax8k.chn
spectrum date:  09-Oct-1993 20:39:14
live time:  5714.00 s
true time:  7200.00 s
num channels:  8192

parameter set:  GeCoax_Pu_120-420 (2019.12.06 14:08)
    Pu Coaxial,  Equilibrium, U235/Pu < 0.2, 120-420keV
    Physical Efficiency,  Gain 0.125 keV/ch, Offset 0 keV
comment:

***************************************************************************
***************************************************************************
diagnostics passed.

                                                   (By Corr)  %Am241/Pu
             Pu238       Pu239      Pu240      Pu241      Pu242      Am241
mass%      0.01149   93.41436    6.38703    0.15674    0.03037    0.16892
sigma      0.00034    0.05703    0.05712    0.00022    0.00067    0.00170
%RSD       2.96%      0.06%      0.89%      0.14%      2.20%      1.01%

%TotPwr    2.59      71.57      17.67      0.21      <  .01       7.66

Specific Power (W/gPu):    (     2.51765 +/-     0.00501)e-003  (  0.20%)

Effective Pu240 fraction:  (     6.46701 +/-     0.05714)e-002  (  0.88%)

Time since chemical separation:     5582.6 +/-   40.4   days   (  0.72%)
```

	Pu238	Pu239
mass%	0.01149	93.41436
sigma	0.00034	0.05703
%RSD	2.96%	0.06%

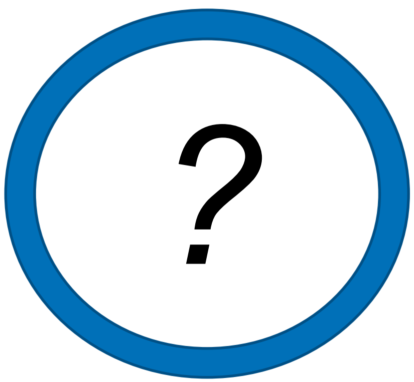
Plutonium Gamma-Ray Spectrum (1 MeV Range)



Plutonium Gamma-Ray Concerns

Gammas only penetrate outer ~1cm of the Pu, we're only measuring the 'skin'

- Which is why calorimetry/neutron is needed, to measure the whole item
- Example: Gamma assay of 4.5kg Pu BeRP ball only gives a few hundred grams



Applications for Gamma Spectroscopy

1) **Finding** material

- Portal monitors
- Source search

2) **Identifying** radionuclides

- Handheld RIIDs
- Manual identification

3) **Quantifying** mass/activity

- Holdup measurements
- Waste measurements

4. Fundamental **science**!

Finding Material: Portal Monitors

Able to **quickly** detect radioactive material

Large, high efficiency

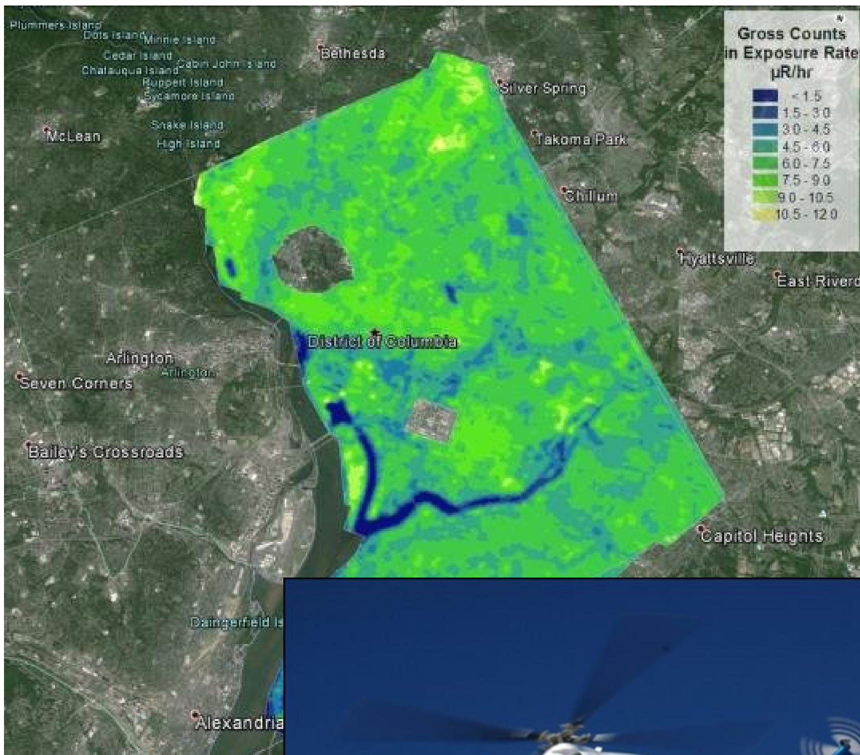
Very poor energy resolution

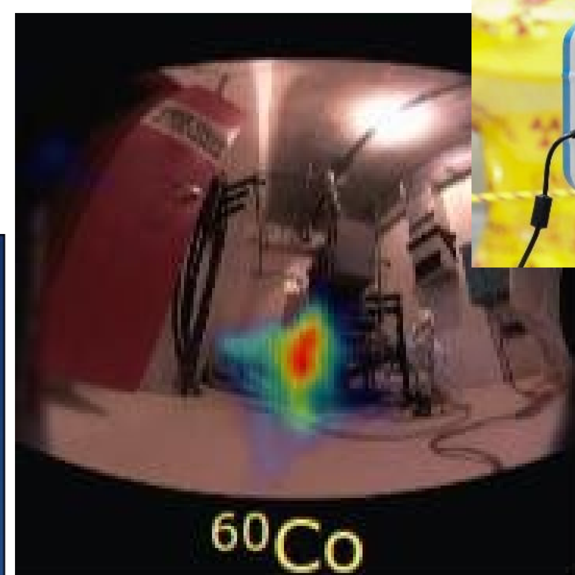
Major focus of NEN-3: International Threat Reduction

Palermo Senator event

Finding Material: Source search

Since gamma-rays are penetrating and passively emitted, they can be used to search for most radioactive materials.





https://h3dgamma.com/

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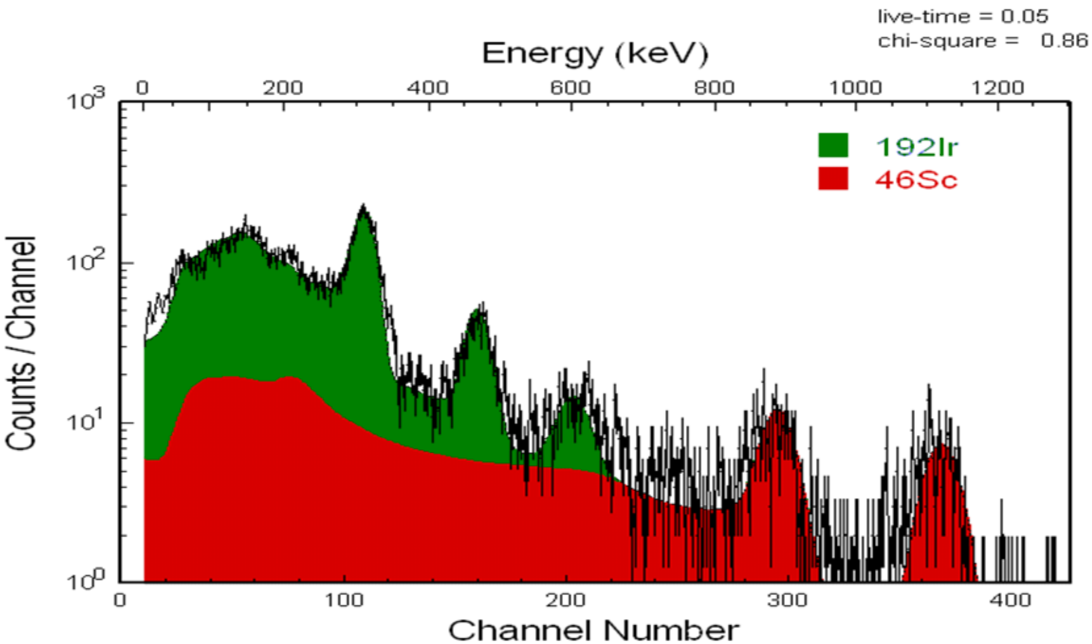
Identifying Material: Handheld RIIDs

Deployable hardware that

1) Collects a spectrum
2) Makes an identification

Usually low resolution, relatively inefficient

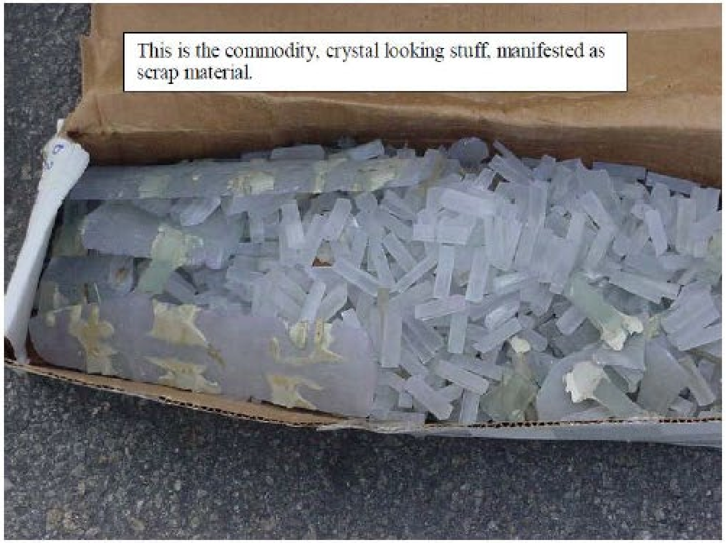


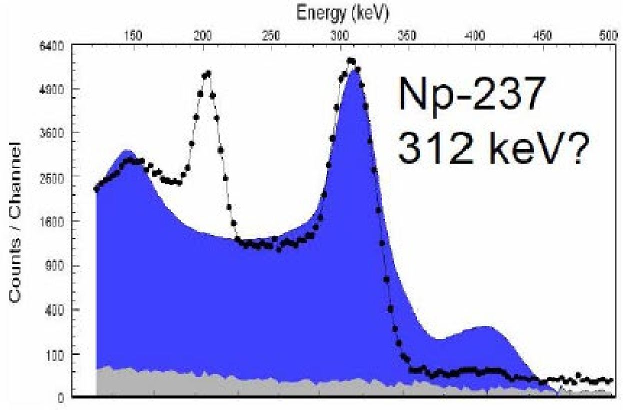
Identifying Material: Manual Identification

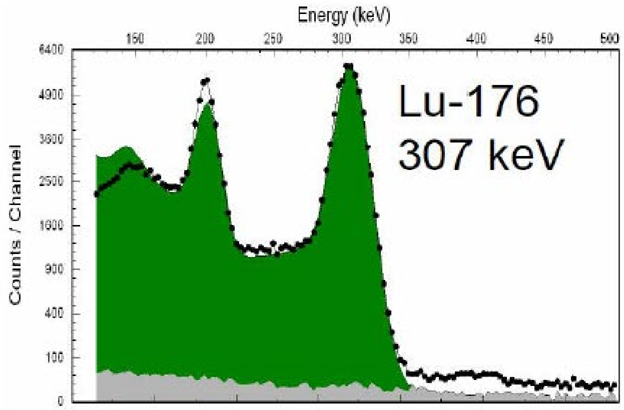
Manual, **human** identification is sometimes needed. Algorithms aren't perfect, and sometimes answers are *alarming*

DOE Radiological Triage provides 24/7 reachback analysis

- **Radioactive boxes contain glass.**
- **GR-135 reports Np-237 & Pu-239.**
- **Triage identifies Lu-176, no Np, no Pu, no threat.**





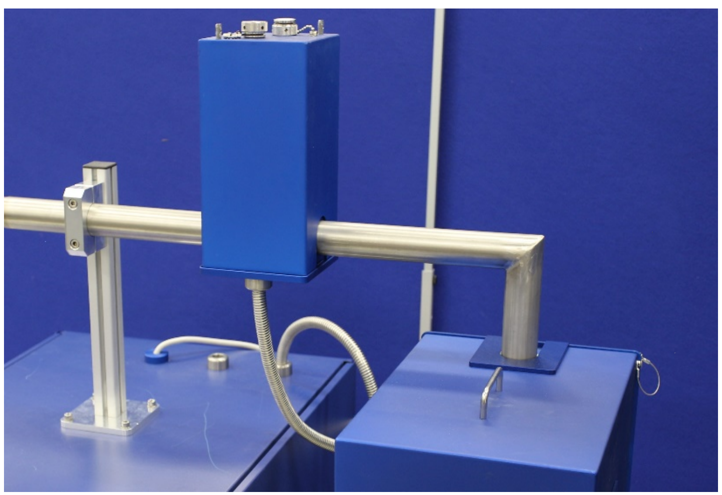


Bonus: Isotopic Analysis

Gamma spectroscopy can be used to determine enrichment of uranium or isotopics of plutonium

NEN-1: Safeguards Science and Technology

- Example: Quantities and enrichment of U-235 limited by international treaties



UF_6 enrichment meter

UF_6 cylinder, high resolution measurement

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Quantifying Material: Waste Measurements

Far-field Gamma Assay:

- Primarily for low-density, homogeneous waste
- Simple analysis

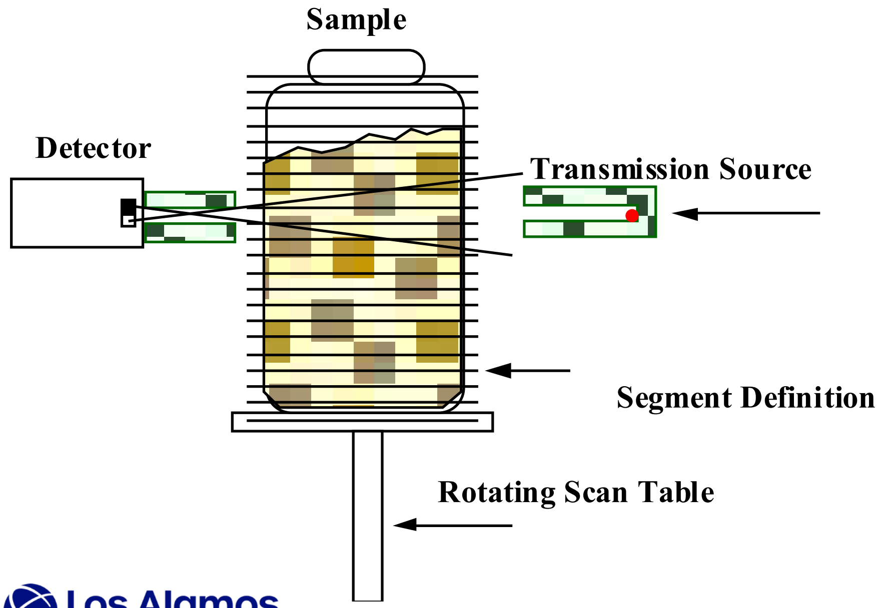
Problem:

- Not all waste is low-density!
- Not all waste is homogeneous!

Quantifying Material: Waste Measurements

Segmented Gamma Scanning

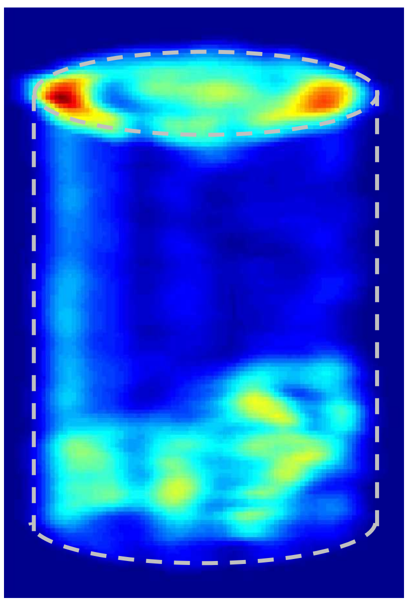
- *Segment* the item into individual layers for the analysis
- Scan along the height of the item while rotating
- Use an external transmission source to account for attenuation



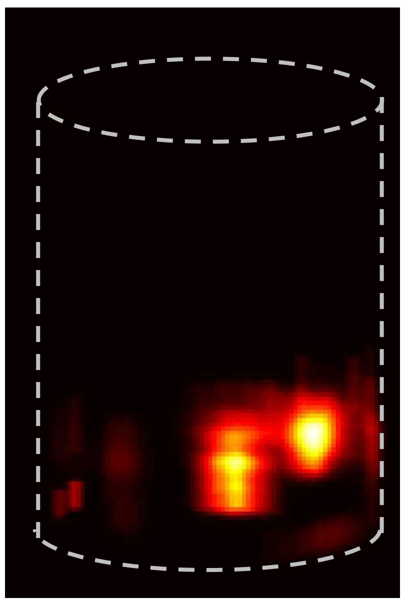
Quantifying Material: Waste Measurements

Tomographic Gamma Scanning

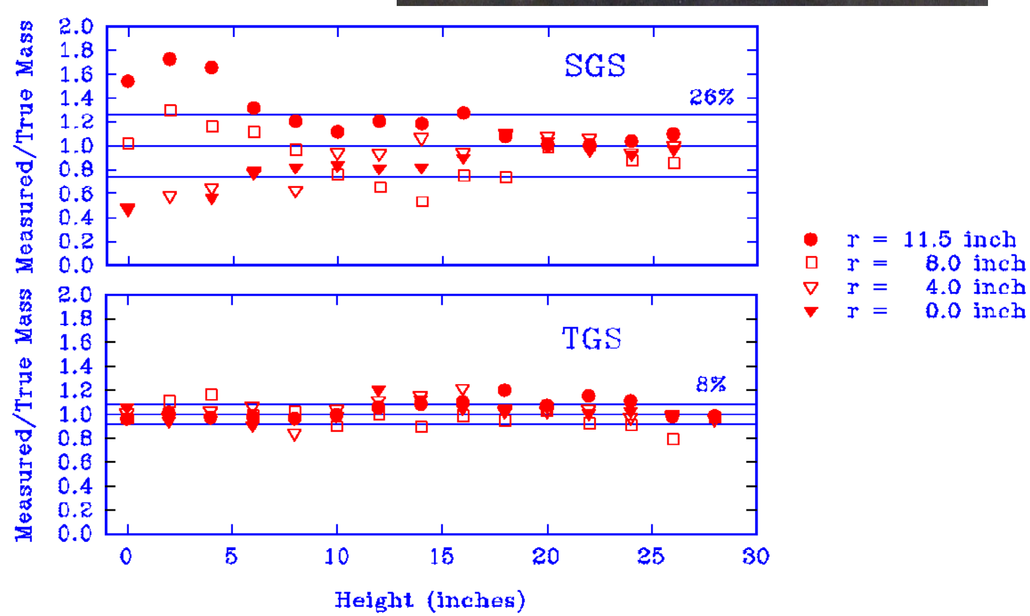
- *Segment* the item into voxels
- Scan along two axes of the item while rotating
- Produce 3D “density map” and “transmission map”



Attenuation



Emission



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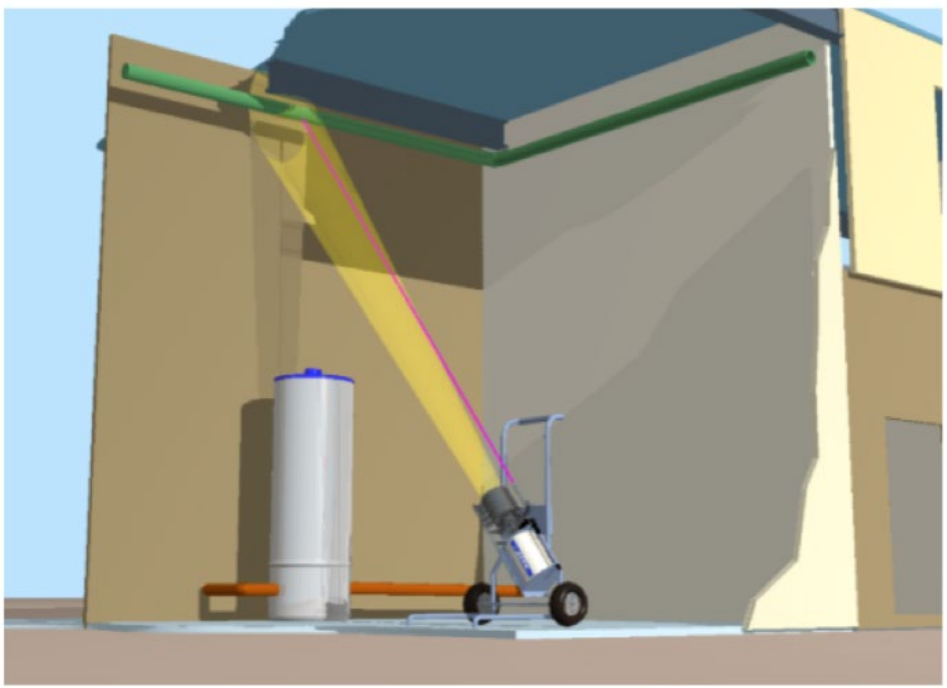
Quantifying Material: Holdup

When processing material, some material is left behind in pipes, blenders, gloveboxes, etc.

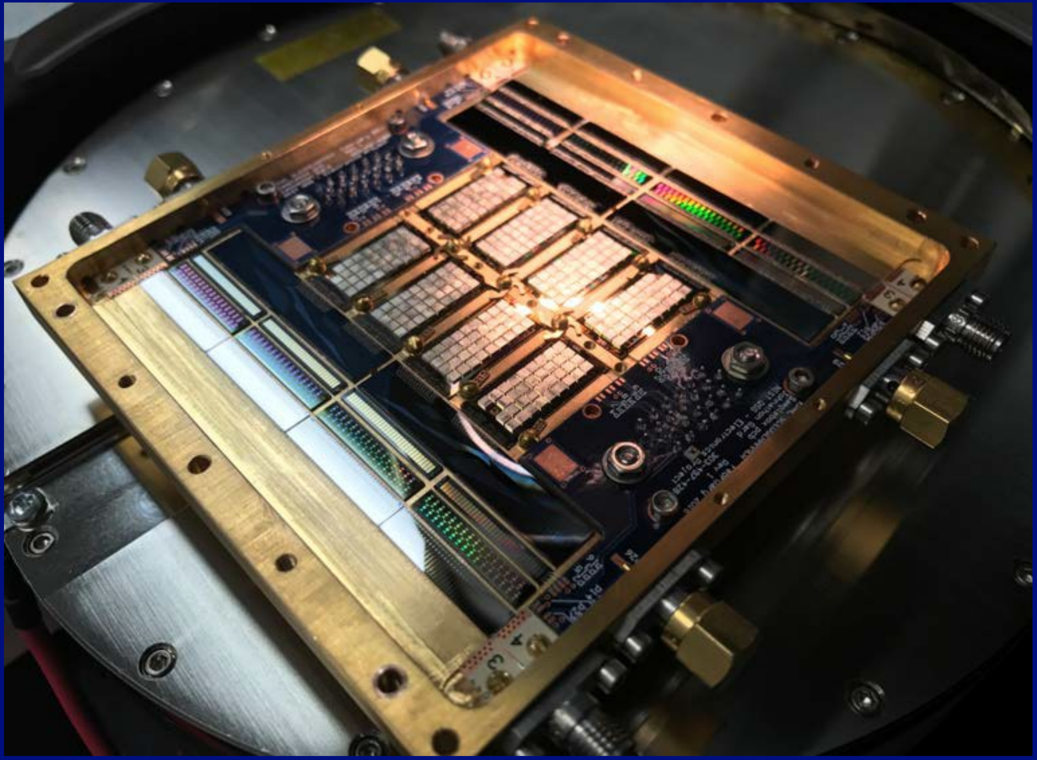
Must be quantified for MC&A, safety, security

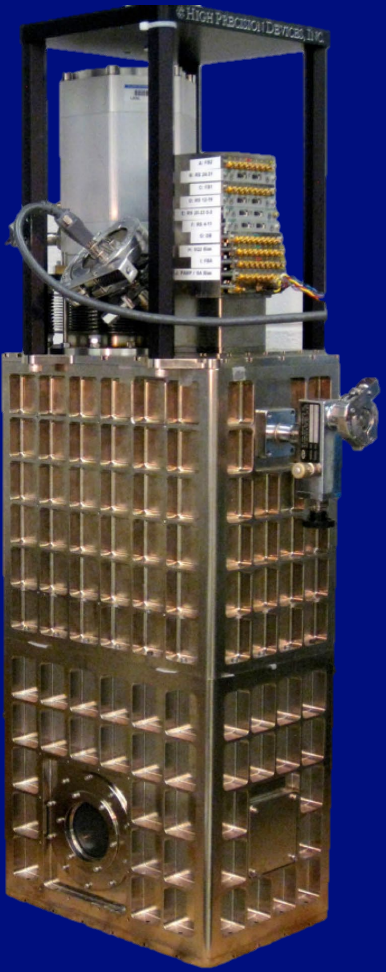


Commercial software:

- ISOCS, ANGLE, ISOTOPIC, SNAP

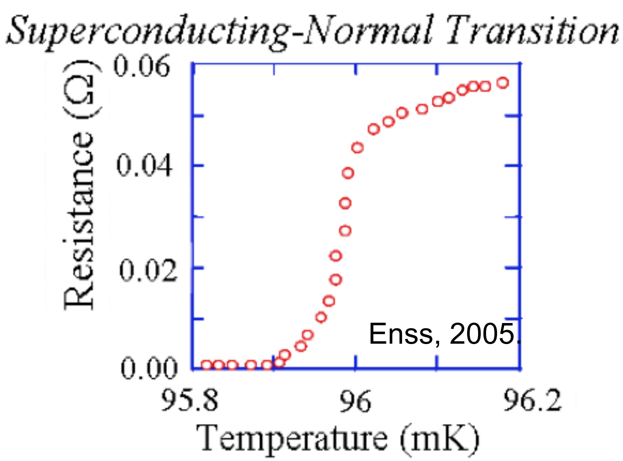
Fundamental science Gamma Spectroscopy R&D: Microcalorimeters

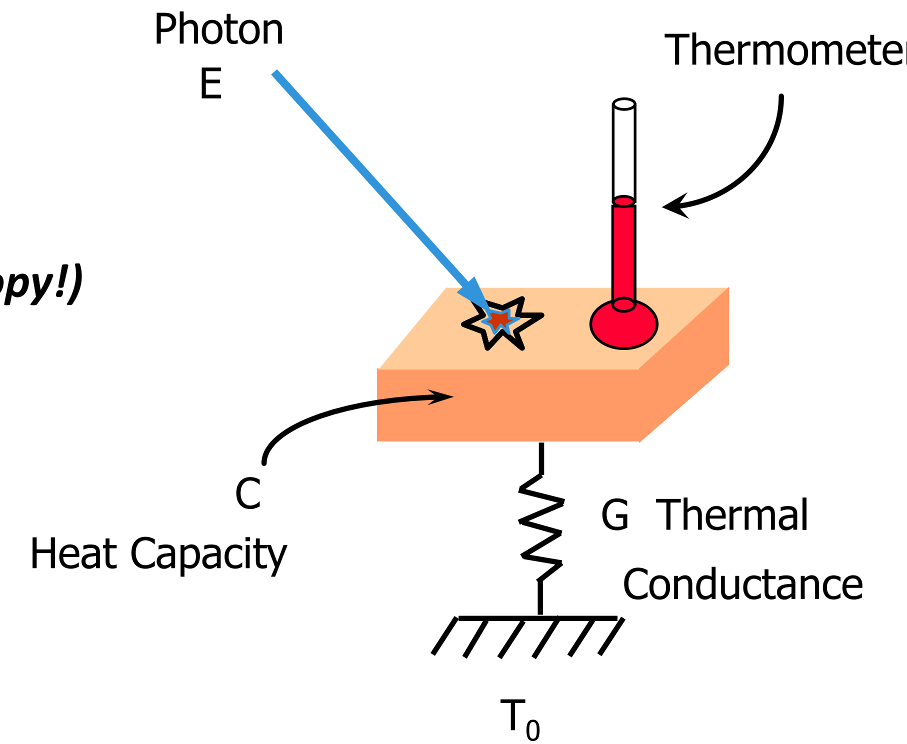


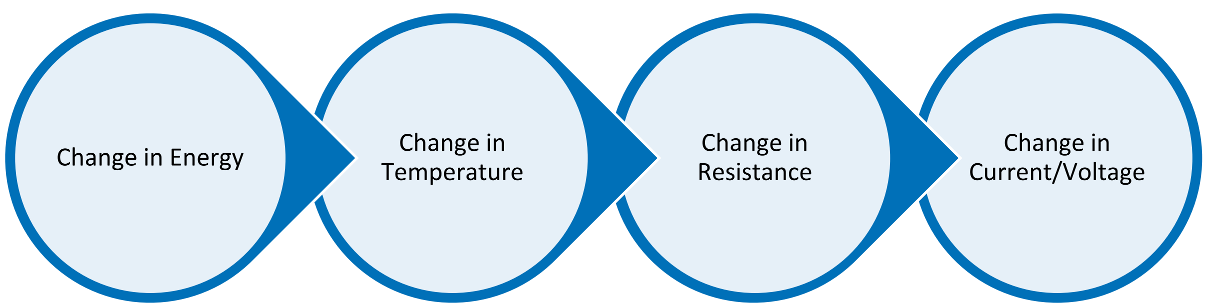


Microcalorimeter

Turn kinetic energy into heat →
Measure with a sensitive thermometer →
Histogram energy from each event (Spectroscopy!)

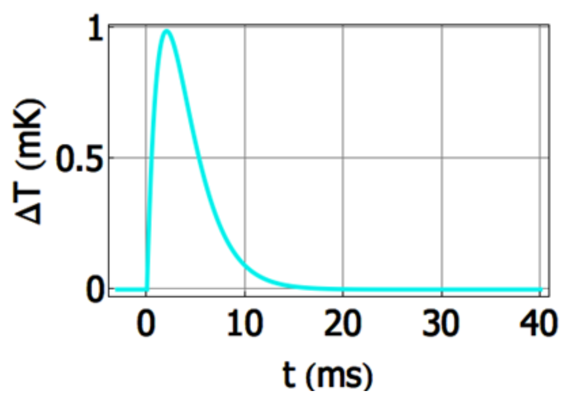






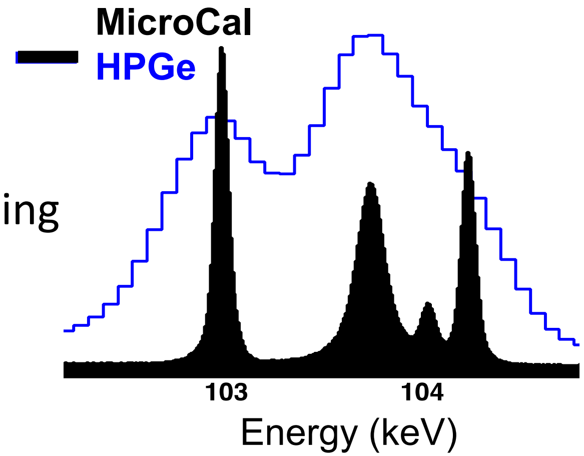
$$E = C \times \Delta T$$

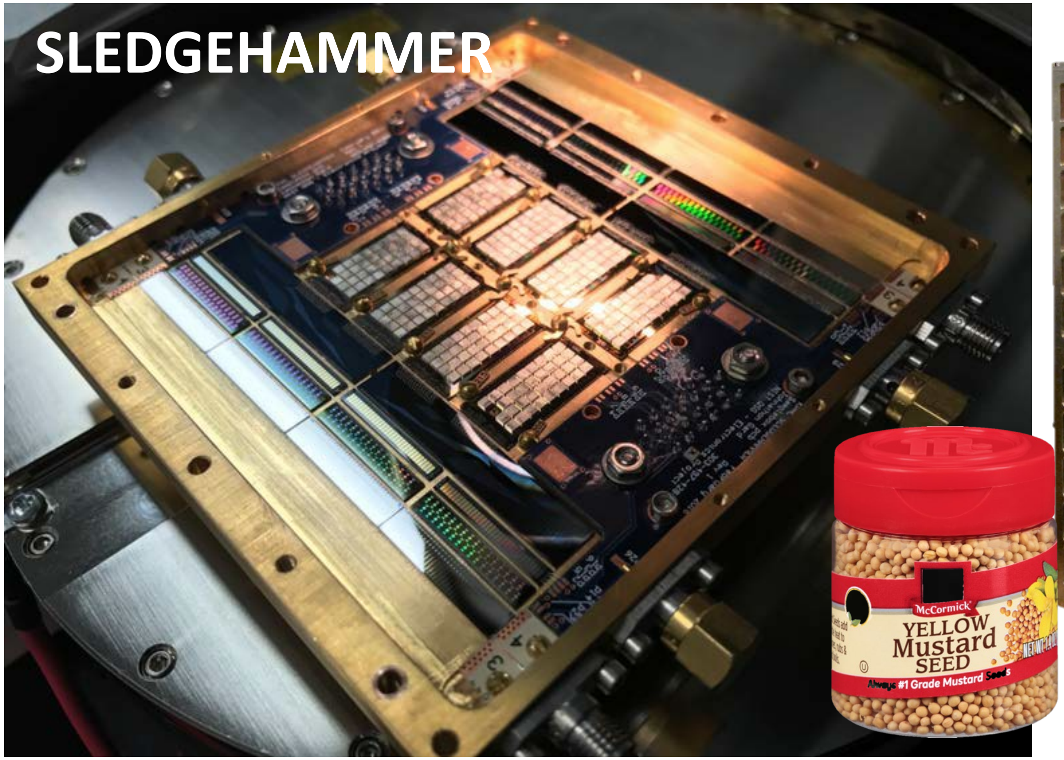
$$V = IR$$

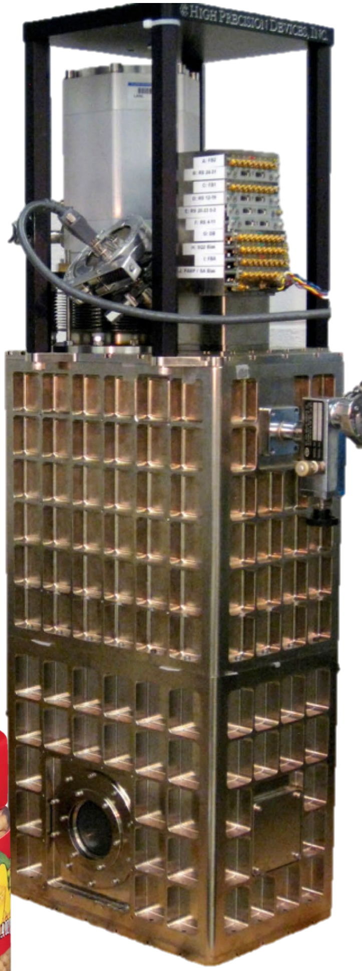


Microcalorimeter

$$\Delta E \approx \sqrt{4k\underset{\text{100 mK}}{T^2}C} \longleftarrow \text{Really small sensing elements}$$

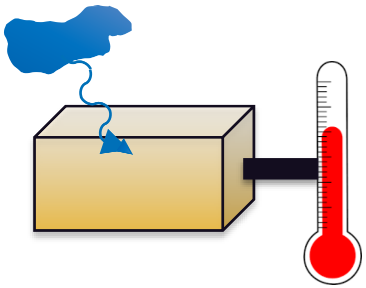


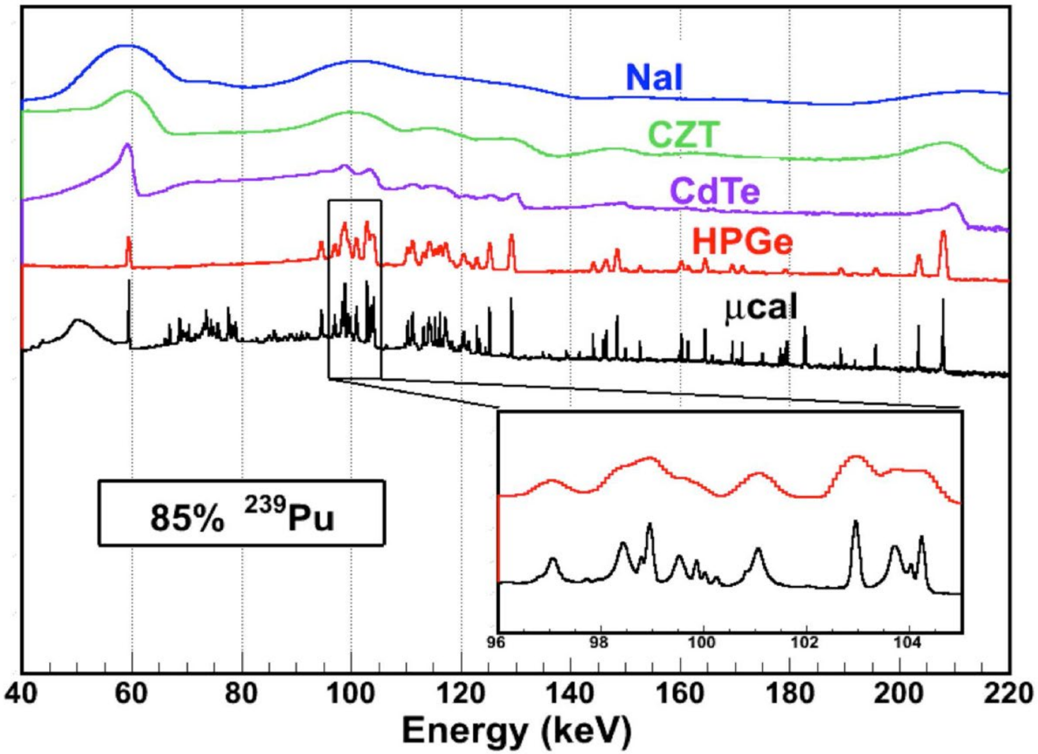




Microcalorimeter

Gamma Ray Spectroscopy





Applications in safeguards and material accounting:

Plutonium isotopic composition

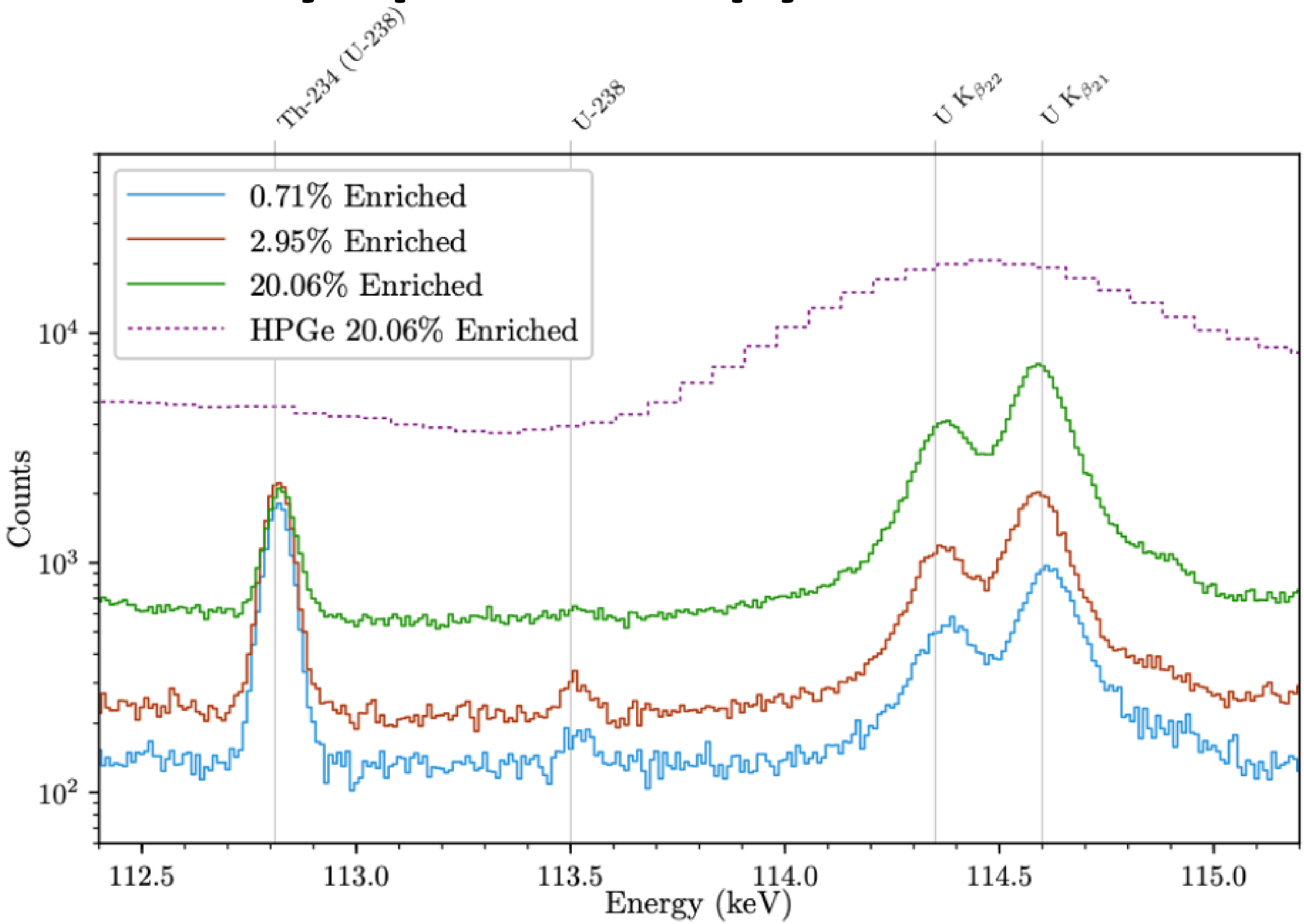
- **Purified U and Pu products**
- **U/TRU products**
- **Fresh MOX fuel**
- **Wastes**

Nuclear data for improved HPGe analysis

- **X-ray line widths**
- **Gamma-ray energies**
- **Gamma-ray branching fractions**

Microcalorimeter

Gamma Ray Spectroscopy



M.P. Croce, arxiv .

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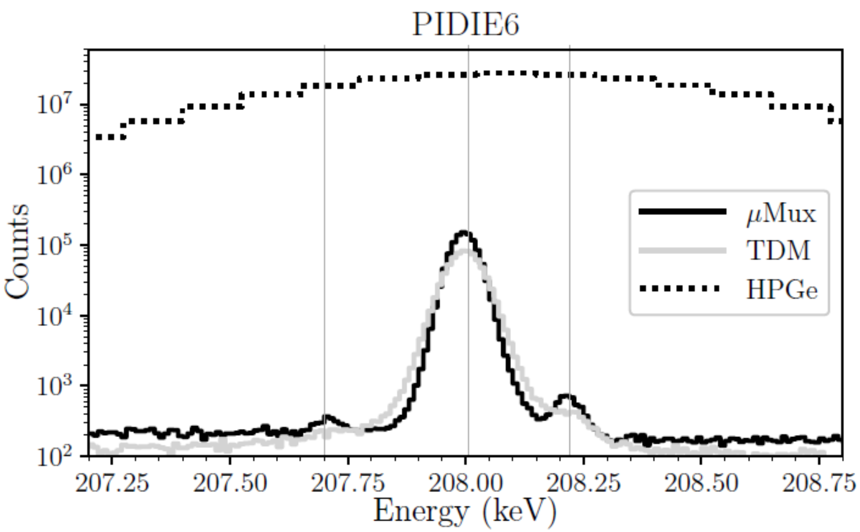
Microcalorimeter

Gamma Ray Spectroscopy



K.E. Koehler, arxiv 2103.15893

Improved nuclear data

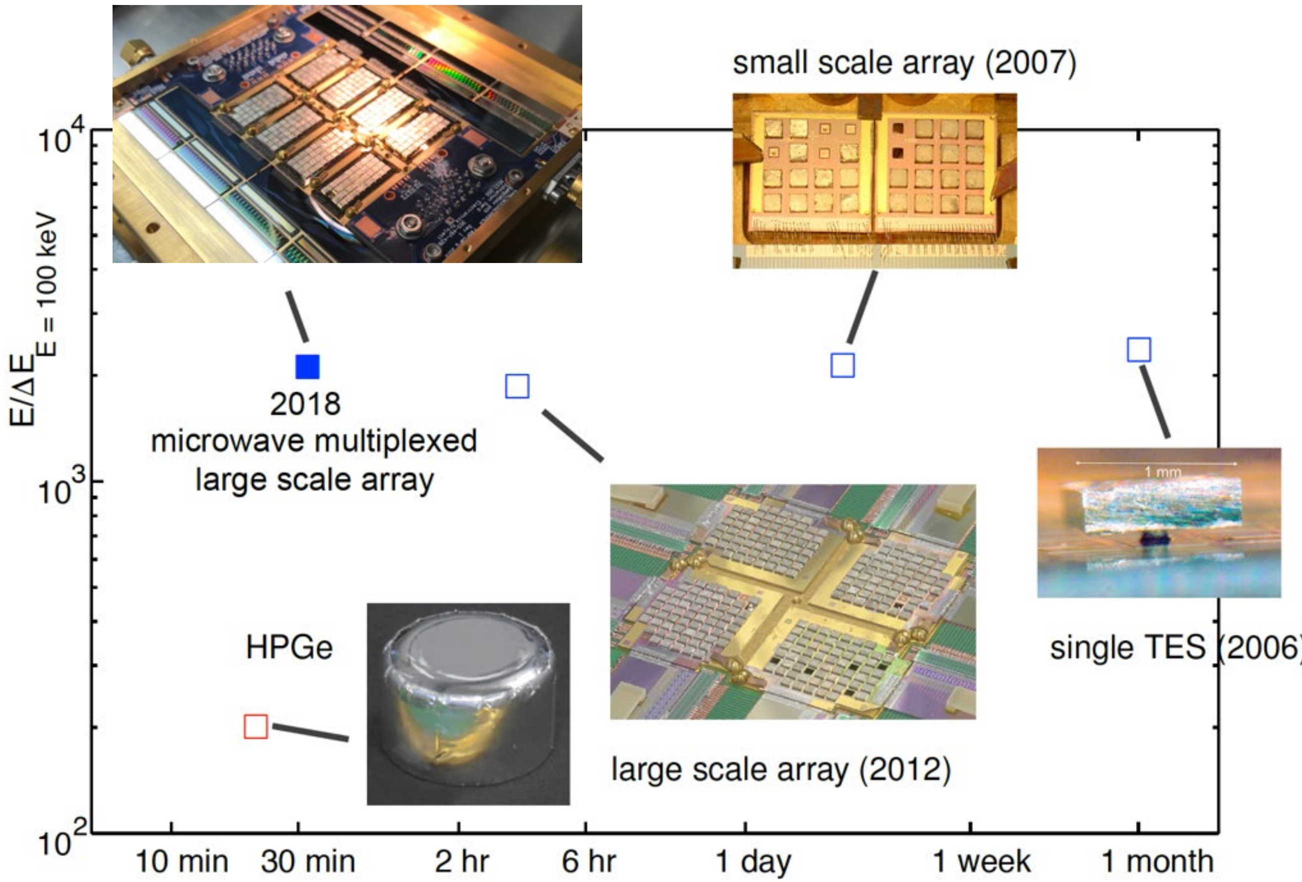
Improved HPGe analysis

Energy [keV]	Isotope	NNDC BR	μ_{BR} [%]	This work BR	μ_{BR} [%]	μ_{BR} Agreement
125.21	^{239}Pu	5.63×10^{-5}	2.7	5.51×10^{-5}	13	−0.2
125.3	^{241}Am	4.08×10^{-3}	2.5	4.08×10^{-3}	1.0	0.0
144.201	^{239}Pu	2.83×10^{-4}	2.1	2.87×10^{-4}	1.0	0.6
146.094	^{239}Pu	1.19×10^{-4}	2.5	1.22×10^{-4}	1.4	0.7
146.55	^{241}Am	4.61×10^{-4}	2.6	4.75×10^{-4}	0.75	1.2
150.04	^{241}Am	7.40×10^{-5}	3.0	7.76×10^{-5}	1.3	1.5
152.72	^{238}Pu	9.29×10^{-4}	0.75	9.46×10^{-4}	0.78	1.7
159.955	^{241}Pu	6.68×10^{-6}	1.1	6.87×10^{-6}	2.0	1.2
160.19	^{239}Pu	6.20×10^{-6}	19	5.82×10^{-7}	331	−2.5
161.45	^{239}Pu	1.23×10^{-4}	1.6	1.20×10^{-4}	1.6	−1.1
161.54	^{241}Am	1.50×10^{-6}	20.0	3.52×10^{-6}	19.9	2.7
164.61	^{241}Pu	4.56×10^{-5}	1.6	4.46×10^{-5}	2.0	−0.9
164.69	^{241}Am	6.67×10^{-5}	3.7	7.78×10^{-5}	4.9	2.4
169.56	^{241}Am	1.73×10^{-4}	2.3	1.72×10^{-4}	0.9	−0.3
171.393	^{239}Pu	1.10×10^{-4}	1.8	1.12×10^{-4}	1.4	0.9
175.07	^{241}Am	1.82×10^{-5}	5.5	1.85×10^{-5}	2.8	0.3
188.23	^{239}Pu	1.09×10^{-5}	10	8.63×10^{-6}	10.8	−1.6
189.36	^{239}Pu	8.30×10^{-5}	1.2	7.91×10^{-5}	1.4	−2.6
191.96	^{241}Am	2.16×10^{-5}	4.6	2.01×10^{-5}	2.8	−1.3
208.005	^{241}Pu	5.19×10^{-4}	1.4	5.34×10^{-4}	1.9	1.2
208.01	^{241}Am	7.91×10^{-4}	2.4	8.08×10^{-4}	5.4	0.4

M.D. Yoho, NIM A, 2020.

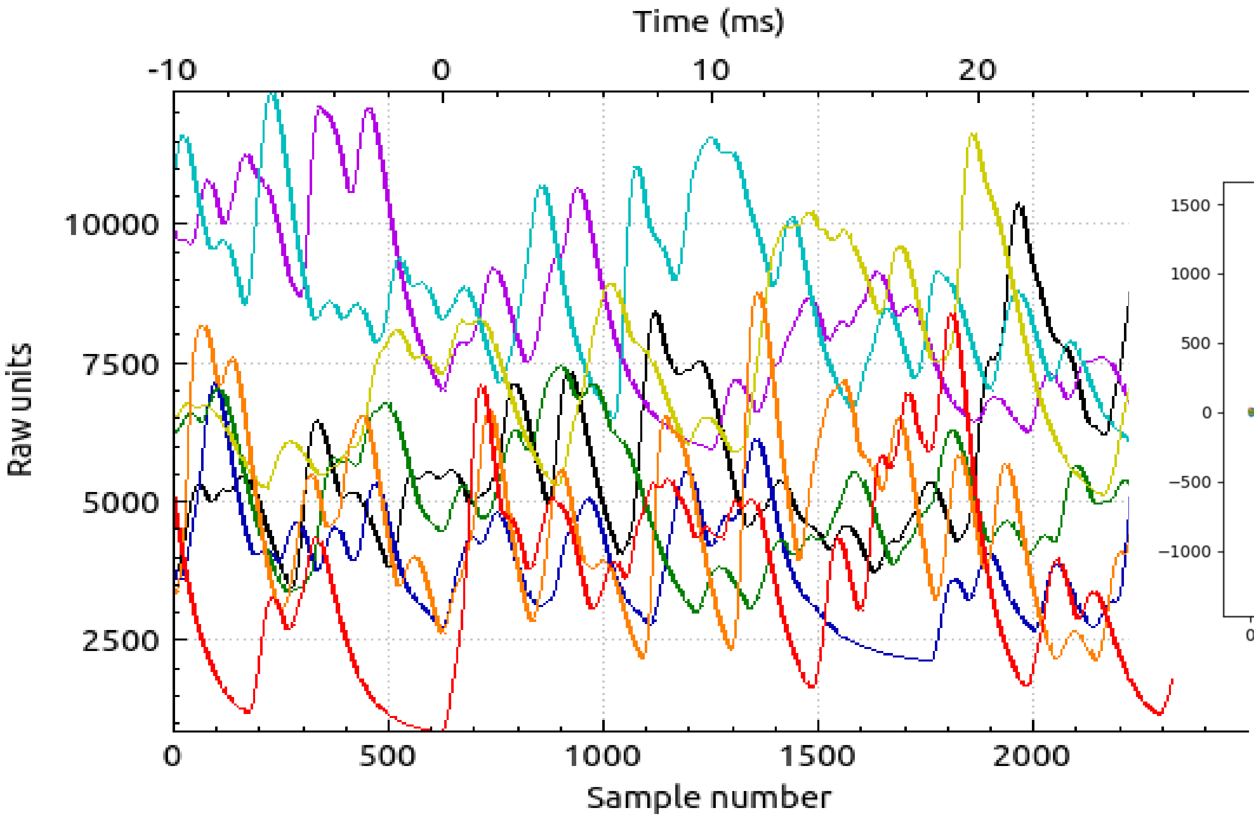
A Bright Future for Microcalorimeters

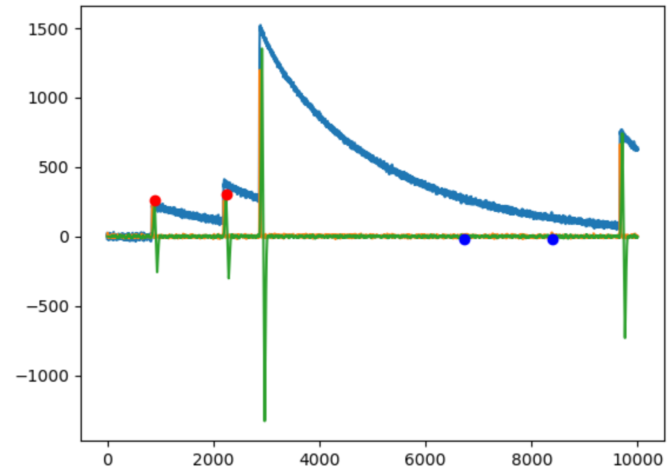
Scalable, high-throughput architecture based on microwave frequency-division multiplexing



resolving power vs. time for quality spectra (5×10^7 counts)

A Bold Future for Microcalorimeters





A realistic future

Stable readout even with extreme pulse pileup

Questions?

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